Dose Assessment for Marine Biota at the Site of Angra dos Reis Nuclear Power Plants, Brazil

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The Piraquara de Fora Bay receives the liquid effluents from the two Brazilian nuclear power plants (NPPs): Angra I that has been operating since 1985 and Angra II that started operating in 2000. The monitoring data set of marine samples over a period of 25 years (from 1985 to 2010) were statistically evaluated. Despite the high presence of nondetect observations, suitable statistical tests were applied to compare ^{60}Co levels in sediments between two periods of time and from different sampling locations. The natural dose and the dose derived from the NPPs routine radioactive releases on the marine biota were assessed by the Erica tool. The highest value of dose rate due to the naturally occurrence radionuclides was estimated to be around 0.6 µGy h^{-1} for phytoplankton, mainly due to internal dose contribution of U^{238}, while fishes received the highest dose (value around 0.4 µGy h^{-1}) due to the radionuclide discharges of the NPPs. Accordingly, the dose rates to the studied species (fish, crustacean, macroalgae, zooplankton and phytoplankton) were clearly below the Erica screening level of 10 µGy h^{-1}, indicating no significant radiological impact of NPPs on these species.

1. INTRODUCTION

The radioprotection principles are presently focused on human protection with the assumption that nonhuman biota would be generally protected if humans were adequately protected. Since this assumption has no unambiguous scientific evidence to support it, efforts have been directed to address evaluation of the effects of ionizing radiation to nonhuman biota by numerous researchers and organizations. Furthermore, an international framework for radiation protection of nonhuman biota has been developed, based on the frameworks being actually adopted in a number of countries as well as the outcome of discussions in international organizations like IAEA and ICRP [1,2]. In fact, in many countries the nuclear facilities need to demonstrate, directly and explicitly, that the biota as a whole is being protected from man-made radionuclides [3,4].

The Nuclear Power Plant site “Central Nuclear Almirante Álvaro Alberto” (CNAAA) is located on the Itaorna Beach, Angra dos Reis, Rio de Janeiro, Brazil (figure 1). The site was designed to have three pressured water reactors: Angra I, which started operation in 1985 and has a nominal capacity of 660 MWe;

Angra II, in commercial operation since 2000, with a nominal capacity of 1350 MWe and Angra III that is under construction. Seawater is used as cooling water for both units: the cooling water is pumped from Itaorna Bay and discharged into Piraquara de Fora Bay (PFB), together with the radioactive liquid effluents from the two power plants. The PFB has an approximate area of 112 km^2, a mean depth of 10 m, and its water temperature has an annual average of 26ºC with a range of 21-29ºC. Permanent water circulation is very slow with moderate hydrodynamism and abundant substrate provided by an irregular coastline and a high rainfall (annual average of 2385 mm)[5].

The environmental radiological assessement of the CNAAA is carried out by the licensee by effluent and environmental monitoring programs. The licensee’s compliance with the requirements is verified by the environmental monitoring control programme carried out by the Institute of Radiation Protection and Dosimetry (IRD/CNEN) [6,7]. The dose evaluation for nonhuman biota is not requested for regulatory purpose.

This paper addresses the evaluation of the dose rate for marine organisms that may be affected by the releasing of the liquid effluents from the Angra I and Angra II nuclear power plants. Data set of 25 years of the monitoring program performed by IRD/CNEN consisting of ^{60}Co, ^{137}Cs, ^{58}Co and ^{54}Mn concentrations in fish, macroalgae, seawater and sediment were evaluated and applied in the Erica toll to calculate dose rates to biota. A comparison of the exposure to marine biota due to radionuclides released by the nuclear power plants with those due to natural radioactivity of the site is also presented.

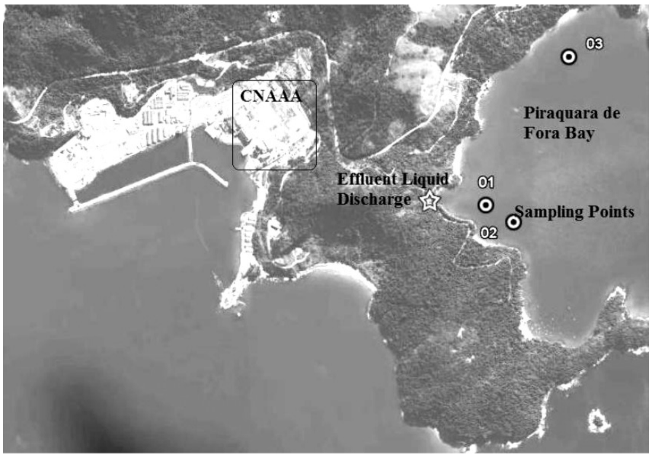

Figure 1. Location of the CNAAA and the sediment sampling points.

2. MATERIAL AND METHODS

Over the past 25 years, the activity concentration in environmental samples from CNAAA site has been monitored, studied and reported by IRD [6,7]. The IRD Laboratories are equipped with state-of-the-art nuclear detectors and counting systems in line with the developments in the area of nuclear measurements. Marine and terrestrial samples are analyzed by gamma spectrometry to evaluate the radionuclide gamma emitters. The radiometric and radiochemical analysis, counting and evaluation of radionuclides are routinely checked by intercomparison exercises. The sequence of analysis for marine organisms (fish and algae) is: i) washing, ii) skinning (fish), iii) oven

drying 100º C , iv) ashing (400º C), v) grounding to a 2mm powder and vi) counting in gamma spectrometer in a suitable geometry. Fresh and dry weights of the samples are recorded. Sediment samples are dried in an oven at 50 °C for at least 20 h, grounding to a 2 mm powder and counting in a suitable geometry. A volume of 5 liters of seawater is transferred to a Marinelli becker before its analysis by gamma spectroscopy. For ^{3}H determination, an aliquot of the distillate seawater is mixed with a scintillation cocktail, and the sample activity is determined by a liquid scintillation spectrometer.

3. RESULTS AND DISCUSSION

Data set of environmental samples of man-made radionuclides usually contains values that are below of the limit of quantification or detection. These data are referred to as minimum detectable activity (MDA), nondetect (ND) or left-censored values [8]. The interpretation of data set including nondetect observations is often a matter of controversy. Due to the lack of the availability of other defensible statistical methods, until recently, the substitution methods (by either the values of DL or DL/2) has been the most used and recommended method to compute the various statistics of interest for the monitoring data sets [9]. Nowadays it has been recommended avoiding the use of DL/2 method even when the percentage of NDs is as low as 5%-10%. Depending on the total number of observations and the frequency of ND observations, a few methods (e.g., Kaplan-Meier and ROS methods) are applicable to estimate summary statistics of data set with multiple detection limit values and data set with detection limits greater than the detected values. Nevertheless, in order to obtain meaningful accurate and statistical results, it is recommended at least to have 10 detected observations [10, 11].

Based on these assumptions, the summaries statistical of the data set from the PFB during the operational period of the power plants were evaluated and the main results are shown in table 1. The number of analyzed and reported samples consists of 57 for Seaweed, 44 for Fish, 40 for Seawater and 40 for each sampling point of sediments. Only a few radionuclides were detected in the marine samples: mainly ^{60}Co in seaweed and sediment, ^{137}Cs in fish and ^{3}H in seawater. For those data sets with more than 10 detected observations, the statistical summary was estimated by the Kaplan-Meir method as suggested by the ProUCL tool for data set with multiple detection values [12](table1).

Table 1. Statistical summary of the main radionuclide concentrations in Fish (Bq/kg $_{\text{fresh weight}}$), Seaweed (Bq/kg $_{\text{dry weight}}$), Seawater (Bq/l) and sediment (Bq/kg dry)

Sample	Radionuclide	Number of Detects	NDs (min:max)	Mean	Max. Observed value
Seaweed	Co-60	16 (28%)	0.23-2.28	1.86	9.20
Fish	Cs-137	16 (36%)	0.04:0.23	0.57	1.76
Seawater	H-3	12 (44%)	5.4:35	21	113
Sediments Pt01+Pt02	Co-60	58 (73%)	0.35:3.60	2.77	16
Sediments Pt01+Pt02	Cs-137	13 (19%)	0.35:3.13	0.82	1.15

In a first approach, the differences between the ^{60}Co levels of the sediment sampling points were tested by the two-sample Gehan´s ranking and Wilcoxon Rank Sum Tests [12]. As there was not provided statistically significant evidence of differences between Pt01 and Pt02 data set, the data were aggregated in a unique population. A time plot of the ^{60}Co levels in sediments of point 01 and 02 is shown in figure 2. In order to avoid confusing between detect and nondetect observations, for ND observations a scatter plot was set using a dashed line from zero to limit detection values [8].

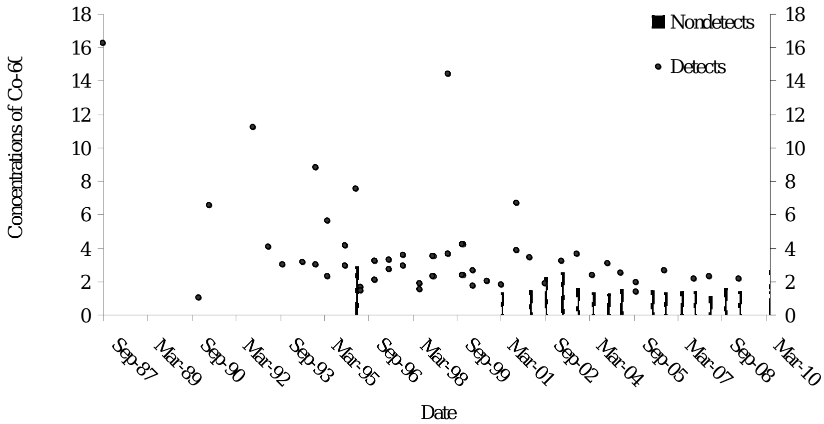

Figure 2. Time plot of the Co-60 concentrations in Pt01 and Pt02 sediments from Piraquara de Fora Bay.

For the sediment from point 03 less than 4 detected observations were collected for all radionuclides. The graphical representation of the temporal levels of ^{60}Co in sediments points out some erratic high values of concentration over the time interval from 1985 to 2000. Since 2000, no value higher than 4 Bq/kg_{dry} was observed (Fig. 2).

Based on the above observation, the two-sample Gehan´s ranking and Wicoxon Rank Sum Tests were applied to test the similarity of the dataset among the sampling points over different periods of time (1985 and 2000) and (2001 and 2010). The outcome clearly indicates that: i) for the first period (1985 to 2000), the ^{60}Co levels in the three sampling points were significantly different and decreases in the following order Pt01>Pt02>PT03, ii) for the second period the ^{60}Co levels in Pt01 were similar with that of Pt03, but both were lower than the levels in Pt02.

Some conclusions can be drawn from the sediments evaluation, there were a decrease in the ^{60}Co levels in the nearest vinicity of the outflow. The increased volume of liquid effluents and in consequence the increased flow of liquid released into PFB caused by the operation of Angra II could have lead to a mobilization, diffusion and dilution of fine sediments. This mobilization may be one of the causes for the decrease in the levels of ^{60}Co in P01 sediments, after the beginning of Angra II operation.

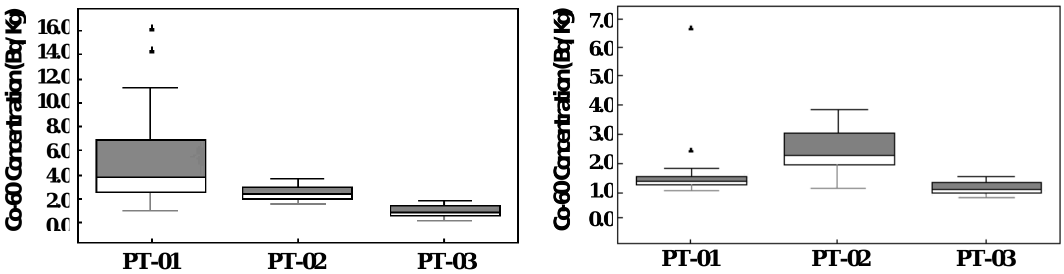

Figure 3. Values of ^{60}Co concentration in the sediment sampling points from 1985 to 2000 (a) and after 2000 (b).

Randomly high concentrations of ^{137}Cs levels in fish and ^{3}H levels in seawater were observed , but no reliable conclusion could be drawn from these data sets.

3.1. **Local Parameters**

Guimarães and Penna-Franca [5] studied the uptake and loss of ^{137}Cs and ^{60}Co by the most abundant green macroalgae in the PFB, *Sargassum filipendula,* and evaluated the sediment distribution coefficients (Kd). These studies were carried out in laboratory experiments using seawater, sediments and algae from PFB (table 2).

Table 2. Concentration factors and distribution coefficients for cesium and cobalt in some marine organisms and sediments from Piraquara de Fora Bay.

Elements	Local Parameters			
	Bioaccumulation factor			Distribution coefficient (Kd)
	Corvine	Shrimp	Seaweed	
Co	130 ± 40	380± 80	554 ±259	50
Cs	41 ±.9	50±10	7±3	4

The estimated bioaccumation factor, expressed on a fresh wt basis, were 7±3 and 554 ±259 for Cs-137 and Co-60 respectively. The factors were normalized for fresh weight by multiplying by 0.16 [13]. The found Kd values were 4 for Cs-137 and 50 for Co-60. These low values of Kd in comparison with the values reported in the literature was imputed to the sandy sediments of PFB. Kamel and Godoy [14] had focused attention on radionuclide accumulation in greatly consumed marine products in the region: a local corvine species (*Micropogonias furnieri*) and shrimp (*Penaeus schimitti*). By determining in situ concentration factors for stable Cs and Co, they had found for corvine a CF value of 41 ±.9 for Cs and of 130 ± 40 for Co, while the CR values for shrimp were 50±10 for Cs and 380± 80 for Co.

3.2. **Background levels of naturally occurring radionuclides**.

Gomes evaluate ^{226}Ra and ^{228}Ra concentrations in seawater to estimate the submarine ground discharge and coastal mixing rates for PFB [15]. The mean concentration in seawater for ^{226}Ra was (1.15±0.53) E-03 Bq/l and for ^{228}Ra (2.11±1.05) E-03 Bq/l. Aiming to study the sedimentation rate and the metal contamination levels, Gomes et al. had taken sediment cores from Ribeira bay,

where the PFB is located. The core slices were analyzed for ^{210}Pb and metals, including U and Th [16]. The mean and standard deviation for ^{210}Pb, ^{238}U and ^{232}Th of the sediments close to PFB were 113±10 Bq/kg, 77±36 Bq/kg and 42±17 Bq/kg, respectively.

3.3. Dose assessment.

The environmental dose rates for marine biota due to the liquid discharge were assessed using Tier 2 ERICA environmental dose tool, Figure 3 [17].

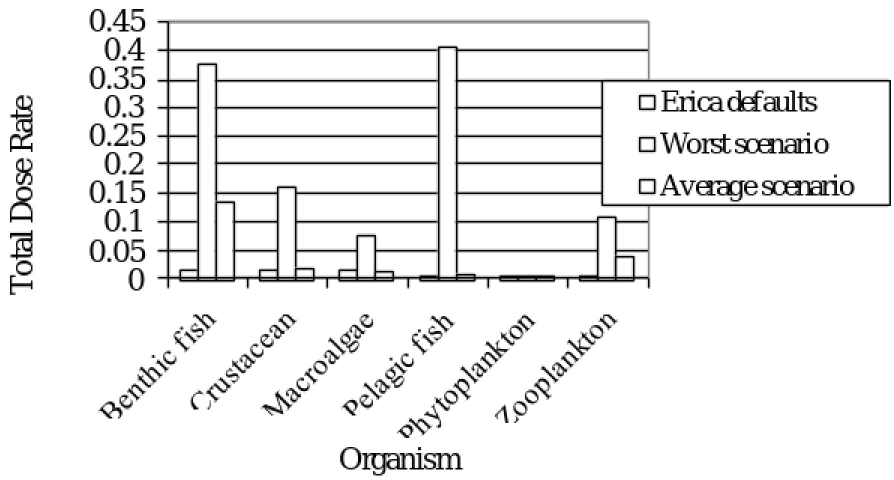

Figure 3. Total dose rate (µGy/h) per marine organism for three scenarios.

The Erica distribution coefficient defaults are 3E05 for Co and 4E03 for Cs, which are much higher than the observed local parameters (table 2). For pelagic fishes and crustaceans the Cs bioaccumulation factors are at the same order of magnitude, whereas the macroalghae Erica default is two order magnitudes higher than the evaluated local one. For ^{60}Co, all bioaccumulation defaults are one order of magnitude higher than the estimated PFB parameters. Given the above data, the highest values of radionuclide concentrations in sediments and seawater with the Erica bioacummulation factor defaults and the locally determinated Kds were used to simulate the worst case scenario for biota. Additionally, the organism total doses were evaluated for an average scenario, a "local" exposure scenario, where the PFB parameters and the mean of ^{60}Co concentrations of the 2001-2010 period (2.8 Bq/kg) were utilized. Also for comparison purpose, a similar scenario to the worst case one was simulated, but using all the default parameters of Erica.

The estimated dose rates in figure 3 show that for all organisms, excepting macroalgae, for the worst case scenario, all dose rate are in the same order of magnitude (10^{-1}). Except for the benthic fish, the dose rate for average and Erica scenarios were one order of magnitude lower than for the worst case scenario. As the external dose has a higher contribution for the total dose than the internal dose, this result emphasizes the key role of the Kd values for the dose evaluation. The fishes were the most exposure organisms (total dose 0.4 µGy/h), nevertheless all the estimated organism doses were lower than the screening value adopted by Erica (10 µGy/h).

The natural background dose rates were in the same order of magnitude of the man-made radionuclide doses for the worst case scenario. Phytoplankton

receives the highest dose rate (0.6 µGy/h), followed by macroalgae (0.2 µGy/h). All the other organisms receive a dose rate in the order of 10^{-2}. Except by Phytoplankton, all organisms' exposures are lower than the minimum typical values presented by ERICA for the same organisms.

4. CONCLUSION

The evaluation of the monitoring data set shows that only a few radionuclides were detected in the marine samples of PFB: mainly ^{60}Co in seaweed and sediment, ^{137}Cs in fish and ^{3}H in seawater. The use of suitable statistical methods allowed to assess varies in ^{60}Co concentration in sediments. They indicate a more elevated concentration of the radionuclide in the nearest vicinity of the effuent discharge and a statistically significant level decreasing between 1985-2000 and the 2000-2010 periods. The ^{60}Co level decreasing may be an outcome of sediment remobilization caused by the increasing of the effluent discharge flow, due to Angra II operation. Dose rates to marine biota under an average and worst case scenarios were clearly below the ERICA screening value of 10 uGy/h, indicating no significant impact of the discharge of radionuclides by the nuclear plants on the considered species. The total exposure of marine biota due to man-made discharged radionuclides is in the same order of magnitude of the natural background exposure.

References

[1] ICRP. ICRP Publication 91. In: Annals of the ICRP, **33**. Pergamon Press Oxford, England (2003)
[2] ICRP. ICRP Publication 103. In: Annals of the ICRP, **37**. Pergamon Press Oxford, England (2007)
[3] Morris R.C. *J.Environ. Radioactiv.***87** (2006)77-100
[4] CNSC-ACRP. Protection of Non-human Biota from Ionizing Radiation.Doc No.INFO-0730. Canadian Nuclear Safety Commission. Ottawa, Canadá (2002)
[5] Guimarães J.R.D. and Penna-Franca E, *Mar. Environ. Res*. **16**, (1985) 77-95.
[6] Martins, N. S. F., Zenaro, R., Nicoli, I.G. and Santos Filho, A. M., XI ENFIR/IV ENAN Joint Nuclear Conference, Rio de Janeiro (1997).
[7] Lauria D.C., Martins N.S.F., Vasconcellos M.L.H., Zenaro R., Peres S.S. and M.A. Pires do Rio M.A. *Appl. Rad. and Isot.* **66** (11)(2008)1636-1638.
[8] Fiévet B. and Vedova C.D. *J.Environ. Radioactiv.***101** (2010)1-7.
[9] USEPA. EPA QA/G-9S.EPA/240/B-06/003. Washington D.C. USA (2006).
[10] Singh, A., Maichle, R., and Lee, S., EPA/600/R-06/022, U.S.A. (2006)
[11] USEPA. ProUCL Version 4.00.05 User Guide (Draft). PA/600/R-07/038 (2010). www.epa.gov.
[12] ProUCL 4.00.05. A Statistical Software. National Exposure Research Lab, EPA, Las Vegas Nevada, USA. (2010).
[13] Vasconcellos L.M.H., Lauria D.C., Silva L.H.C. and Taddei J.F, *Química Nova*, **22** (6) (1999)889-893.
[14] Kamel L.N., Determinação de Fatores de Concentração para cromo, cesio, ferro e cobalto em corvina e camarão nas circunvizinhanças da central nuclear Almirante Alvaro Alberto (CNAAA). Centro Biomédico da universidade do Estado do Rio de Janeiro. UERJ. 1988. 128pp.
[15] Gomes F.C., Godoy J.M., Carvalho Z.L. and Lopes R.T., *Radioprotection*. **44**, 5 (2009) 237-241.

[16] Gomes F.C., Godoy J.M., Godoy M.L, Lara de Carvalho Z, Tadeu Lopes R, Sanchez-Cabeza JA, Drude de Lacerda L, Cesar Wasserman J. *Mar Pollut Bull*. **59**(4-7) (2009)123-133.
[17] Brown , J.E., Afonso, B., Avila R., Beresford, Copplestone D., Prohl G., Ulanovsky A. *J.Environ. Radioactiv*.**99** (2008)1371-1383.